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Original Article

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Drying Kinetics of *Cecropia pachystachya* Leaves

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ABSTRACT

The species *Cecropia pachystachya* has important medicinal purposes and its leaves have been used in pharmaceutical research, so the drying of this product may help maintaining its chemical properties and ensure safe storage. Thus, the objective of this study was to select mathematical models to represent the drying kinetics of *Cecropia pachystachya* leaves, determine the effective diffusion coefficient and obtain the activation energy during drying at different temperatures. Leaves were dried in an oven under five temperature conditions (40, 50, 60 and 70 °C), until reaching hygroscopic equilibrium moisture content. Among the models analyzed, the Logarithmic model best represented the drying kinetics at temperatures of 40 and 60 °C, whereas Modified Henderson & Pabis and Dryceleaves represented temperatures of 50 and 70 °C, respectively. The effective diffusion coefficient increased with increasing air temperature, and the activation energy for liquid diffusion in the drying process was 64.53 kJ mol-1.

Keywords: activation energy, embaúba, liquid diffusion, mathematical models, technology of forest products.

1. INTRODUCTION

Cecropia pachystachya, popularly known as 'embaúba' in Brazil (Costa et al., 2011), is a medium size, pioneer tree of the Urticaceae family, with height from 4 to 8 meters. It prefers shaded, humid sites and has simple alternate leaves with 8 parts of 40 cm, on average (Salman et al., 2008), with fast growth. Several studies using the Urticaceae family have been carried out due to its diversity of more than 2000 species and multiple medicinal uses. *C. pachystachya* became even more important for research due to the recent full characterization of its chloroplast DNA performed by Wu et al. (2017), which facilitates its use in studies related to the Urticaceae family, being very relevant in the most diverse areas of biology and medicine.

Five species of the genus occur in Brazil: *Cecropia glaziou* Sneth, *C. hololeuca* Miq, *C. pachystachya* Trécul, *C. purpurascens* Berg and *C. sciadophylla* Mart. *C. pachystachya*, popularly known as 'embaúba', which may reach 7 m in height with trunk diameter ranging from 15 to 25 cm (Bocchese et al., 2008).

C. pachystachya leaves have been widely studied aiming at new pharmaceutical products intended for the treatment of several diseases, through their antipathogenic compounds (Brango-Vanegas et al., 2014; Souza et al., 2014), functions such as antidepressant and protection from oxidative stress (Ortmann et al., 2016), anti-inflammatory and antioxidant, which can be attributed to the presence of phenolic compounds (Pacheco et al., 2014), more specifically flavonoids (Talhi & Silva, 2012).

Drying of products with medicinal and pharmacological potential aims, among other aspects, to prepare them for safe storage, reduce enzymatic degradation, maintain chemical properties and operationalize their use in the industrial production with volume reduction (Goneli et al., 2014; Martins et al., 2015; Gasparin et al., 2017). Drying is also known as a process that extends the consumption period of plant materials (Horuz et al., 2017).

Various drying conditions should be tested to adjust the characteristics of each product during moisture content reduction, and theoretical mathematical models have been constantly used in literature to predict this phenomenon (Silva et al., 2017a; Maciel et al., 2017; Sonmete et al., 2017).

Given the above, this study aimed to select mathematical models capable of representing the drying kinetics of *C. pachystachya* leaves, as well as to determine and evaluate the effective diffusion coefficient, in addition to obtaining the activation energy for the drying process at different air temperatures.

2. MATERIAL AND METHODS

The experiment was conducted at the Laboratory of Post-harvest of Plant Products of the Federal Institute of Goiás – Campus of Rio Verde, using *C. pachystachya* ('embaúba') leaves collected from trees located in the preservation area of the campus at coordinates 17°48'3.52"S, 50°54'27.33"W, and mean altitude of 720.0 m a.s.l., deposited in the herbarium of the Federal Institute of Goiás – Campus of Rio Verde under number 1009 and identified by specialist PhD. André Luiz Gagliote.

Leaves were collected from the third middle of trees, between 6 and 7 a.m., time of maximum leaf turgor, and stored in plastic bags full of CO_2, in order to inhibit water loss during transportation from the collection site to the processing laboratory. The plant material was subjected to cleaning and weighing prior to drying, using an analytical scale, with 0.01 g resolution, determining the wet weight of samples and weight of containers. Containers consisted of perforated metal trays with diameter of 28.0 cm.

Three leaves per replicate were used due to the large leaf area of the species, with 3 replicates per temperature condition during drying in oven with forced air circulation, regulated at 40, 50, 60 and 70 °C.

The gravimetric method was used to reduce the moisture content of *C. pachystachya*, with periodical weighing until hygroscopic equilibrium, when constant weight was achieved during the drying process. Before and after drying, moisture contents were determined by the method recommended by ASAE (2000), for fodder and leaves, in oven with forced air circulation at 103 ± 1 °C, for 24 hours. Room air temperature and relative humidity were monitored using a datalogger and the average relative humidity (RH%) inside the oven during the drying process was estimated by the GRAPSI v.8 software (Melo et al., 2004).

Experimental data were used to determine the moisture content ratios (RX) using Equation 1 (Sharaf-Eldeen et al., 1980).

$$RX = \frac{X^* - X_e^*}{X_i^* - X_e^*} \quad (1)$$

where: RX = moisture content ratio of the product, dimensionless; X* = moisture content of the product, decimal (d.b.); X_i* = initial moisture content of the product, decimal (d.b.); and X_e* = equilibrium moisture content of the product, decimal (d.b.).

Then, mathematical models commonly used in literature to represent the drying kinetics of agricultural products, as well as the model proposed in the present study, called Dryceleaves (Drying of Cecropia leaves), were fitted to data, as described in Table 1.

Models were fitted by nonlinear regression analysis using the Gauss-Newton method. Models were selected considering the magnitude of the following coefficients: determination (R^2), mean relative error (P) (Equation 15) and mean estimated error (SE) (Equation 16), according to Smaniotto et al. (2017). For P, value ≤ 10% was considered as the main criterion to select the models, as well established in studies related to the drying of biological products.

$$P = \frac{100}{N}\Sigma\frac{|Y - \hat{Y}|}{Y} \quad (15)$$

$$SE = \sqrt{\frac{\sum(Y - \hat{Y})^2}{DF}} \quad (16)$$

where: Y = experimental value; Ŷ = value estimated by the model; N = number of experimental observations; DF = degrees of freedom of the model (difference between number of observations and number of parameters of the model).

Akaike's Information Criterion (AIC) and Schwarz's Bayesian Information Criterion (BIC), represented by Equations 17 and 18 (Burnham & Anderson, 2004), respectively, were used as complementary and discriminating indicators. These indices were calculated by the R statistical program, so that the lower the values found, the better the fits of the model used in the study.

Table 1. Mathematical models used to predict the drying phenomenon of agricultural products.

Model designation	Model	Equations	References
Wang & Singh	$RX = 1 + a.t + b.t^2$	(2)	Moyne et al. (1992)
Verma	$RX = a.\exp(-k.t) + (1-a)\exp(-k1.t)$	(3)	Verma et al. (1985)
Thompson	$RX = \exp\left\{\left[-a - (a2 + 4.b.t)^{0.5}\right]/2.b\right\}$	(4)	Thompson et al. (1968)
Page	$RX = \exp(-k.t^n)$	(5)	Agrawal & Singh (1978)
Newton	$RX = \exp(-k.t)$	(6)	O'Callaghan et al. (1971)
Midilli	$RX = a.\exp(-k.t^n) + b.t$	(7)	Arslan & Özcan (2008)
Logarithmic	$RX = a.\exp(-k.t) + c$	(8)	Yagcioglu et al. (1999)
Henderson & Pabis	$RX = a.\exp(-k.t)$	(9)	Henderson (1974)
Modified Henderson & Pabis	$RX = a.\exp(-k.t) + b.\exp(-ko.t) + c.\exp(-k1.t)$	(10)	Karathanos (1999)
Two-term exponential	$RX = a.\exp(-k.t) + (1-a)\exp(-k.a.t)$	(11)	Sharaf-Eldeen et al. (1980)
Two terms	$RX = a.\exp(-ko.t) + b.\exp(-k1.t)$	(12)	Henderson (1974)
Approximation of Diffusion	$RX = a.\exp(-k.t) + (1-a)\exp(-k.b.t)$	(13)	Kassem (1998)
Dryceleaves (model proposed)	$RX = a + b.t^{2.5} + c.\exp(-t)$	(14)	

t = drying time; h; k, k_o, k_1 = drying constants, h^{-1}; a, b, c, n = model coefficients; Eq. = equation.

$$AIC = -2\log L + 2(p) \quad (17)$$

$$BIC = -2\log L + p\log(N - r) \quad (18)$$

where: p = number of model parameters to be estimated; N = total number of observations; r = rank of matrix X (incidence matrix for fixed effects); and L = maximum likelihood estimator of error variance.

The effective diffusion coefficient for *C. pachystachya* leaves was obtained by means of the Infinite Slab model, with approximation of 8 terms, as represented in Equation 19 (Smaniotto et al., 2017).

$$RX = \frac{X^* - X_e^*}{X_i^* - X_e^*} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left[-\frac{(2n+1)^2 . \pi^2 . D.t}{4} . \left(\frac{S}{V}\right)^2\right] \quad (19)$$

where: RX = moisture content ratio of the product, dimensionless; D = effective diffusion coefficient, $m^2\ s^{-1}$; n = number of terms; S = surface area of the product, m^2; and V = leaf volume, m^3.

Surface area was determined using the ImageJ.® software (Image Processing and Analysis in Java), which consists in an image integrator. Images were previously obtained by photographing the plant material on a white background of known scale. Leaf volume was determined considering the surface area and leaf thickness, measured using a digital caliper. The average surface area of leaves used was 1.45 x 10^{-1} m^2, with thickness of 4.60 x 10^{-4} m and average volume of 6.66 x 10^{-5} m^3.

The Arrhenius expression describes the ratio between diffusion coefficient (D) and the variation in drying temperature according to the following expression.

$$D = D_o . \exp\left(\frac{E_a}{R.T_a}\right) \quad (20)$$

where: D = liquid diffusion coefficient, $m^2\ s^{-1}$; D_o = pre-exponential factor; E_a = activation energy, kJ mol^{-1}; R = universal gas constant, equal to 8.314 kJ $Kmol^{-1}$; and T_a = absolute temperature, K.

3. RESULTS AND DISCUSSION

Reduction in the moisture content of leaves occurred within the range from 0.0017 to 0.0212 (dry basis), with drying times of 31, 19, 8 and 2 hours for temperatures of 40, 50, 60 and 70 °C. Under these conditions, RH% values estimated inside the oven were 24.84% (40 °C), 14.85% (50 °C), 7.79 (60 °C) and 4.93% (70 °C).

In the drying process, the elevation of air temperature increases the speed with which water is removed from the material and, for Gomes et al. (2017), this phenomenon is due to the increase in the difference of saturated air vapor pressure inside the plant product, resulting in water movement from inside the leaf to the drying air in a shorter period of time. This behavior has been reported in various studies, such as those conducted by Sahin & Öztürk (2016) with fig fruits, Smaniotto et al. (2017) with sunflower grains, Horuz et al. (2017) with apricot fruits and Mghazli et al. (2017) with rosemary leaves.

Based on the mean relative error (P<10%) (Table 2), being an eliminatory statistical parameter, theoretical models with the lowest magnitude at temperature of

Table 2. Mean relative error (P) and determination coefficient (R^2) of mathematical models fitted in the drying of *C. pachystachya* leaves under different temperature conditions.

Models	40 °C		50 °C		60 °C		70 °C	
	P	R^2	P	R^2	P	R^2	P	R^2
Approximation of diffusion	7.47	0.997	31.82	0.999	10.96	0.996	329.36	0.991
Two terms	8.75	0.998	52.29	0.981	11.25	0.996	53.52	0.999
Two-term Exponential	13.57	0.997	53.48	0.980	20.02	0.995	329.35	0.995
Henderson & Pabis	21.60	0.997	52.29	0.981	23.65	0.995	319.13	0.992
Mod. Henderson & Pabis	21.60	0.997	6.84	1.000	84.72	0.701	53.47	0.999
Logarithmic	7.93	0.998	119.96	0.992	9.94	0.996	56.07	0.999
Newton	24.65	0.995	53.48	0.980	23.88	0.995	329.35	0.991
Page	16.33	0.997	33.86	0.990	22.38	0.995	213.88	0.995
Thompson	11.71	0.997	21.22	0.994	19.81	0.995	329.41	0.991
Verma	7.48	0.997	31.81	0.998	10.96	0.996	54.48	0.999
Wang & Singh	90.26	0.922	680.33	0.399	125.40	0.922	176.83	0.923
Midilli	10.86	0.998	75.54	0.992	12.93	0.997	85.30	0.999
Dryceleaves	116.15	0.795	121.13	0.984	17.24	0.990	3.16	0.998

40 °C were Approximation of diffusion, Two terms, Logarithmic and Verma, whereas Modified Henderson & Pabis, Logarithmic and Dryceleaves proved to be efficient at 50, 60 and 70 °C, respectively. Thus, considering this parameter, only one model was fitted for the conditions of drying temperatures, except for 40 °C. Satisfactory mean relative errors at 40 °C have also been found in the drying of lemon balm using the Approximation of diffusion model (Barbosa et al., 2007) and 'timbó' (*Serjania marginata* Casar) using the Logarithmic model (Martins et al., 2015).

Determination coefficients were higher than 0.95, except for the Modified Henderson & Pabis model at temperature of 60 °C, Wang & Singh at all drying temperatures and the Dryceleaves model at 40 °C (Table 2). Although most of these models under the drying conditions of this study resulted in high R^2 values, this coefficient alone is not determinant for the choice of nonlinear models fitted in the drying of *C. pachystachya* leaves. Complementary analyses with other parameters are necessary, as those used in studies with different plant materials and drying conditions (Darvishi et al., 2014; Camicia et al., 2015; Rosa et al., 2017; Moscon et al., 2017).

For the mean estimated error (SE), according to the selection of models by the P criterion, the Logarithmic model fitted best at temperatures of 40 and 60 °C, with values of 1.0 x 10^{-4} and 1.8 x 10^{-4}. For the other temperature conditions, SE values were equal to 0.2 x 10^{-4} for Modified Henderson & Pabis at 50 °C and 0.8 x 10^{-4} for the Dryceleaves model at temperature of 70 °C (Table 3).

Considering P and SE, the Logarithmic model was the one that best represented the drying kinetics of *Solanum lycocarpum* A. St.-Hil leaves at temperatures of 40, 50 and 60 °C (Reis et al., 2012) and the Modified Henderson & Pabis was the best for *Schinus terebinthifolius* Raddi at temperatures of 40, 50, 60 and 70 °C (Goneli et al., 2014), corroborating the present study, in which the drying kinetics was satisfactorily represented by the Logarithmic model at temperatures of 40 and 60 °C, and by the Modified Henderson & Pabis model at temperature of 50 °C.

For Van Boekel (2008), the discrimination of models should be parsimonious and, when several models have reasonable fits, criteria such as Akaike's and Schwarz's Bayesian become useful tools to select the most efficient to predict a certain behavior. Thus, the AIC and BIC criteria were applied as a method to discriminate the most efficient model to represent the drying process of C. *pachystachya* at temperature of 40 °C, since several models have P<10%.

According to Table 4, the Logarithmic model resulted in lower magnitude for AIC and BIC criteria, corroborating results found for the mean relative error. In addition, among the most adequate models for drying at 40 °C, this as the lowest number of parameters and is recommended to represent moisture content reduction in *C. pachystachya* at this temperature. Following the classification order, the next models were Two terms, Verma and Approximation of diffusion.

The models with the best fits, suggested in the present study, are graphically represented in the drying curves

Table 3. Mean estimated error (SE) of mathematical models fitted in the drying of *C. pachystachya* leaves under different temperature conditions.

Models	40 °C	50 °C	60 °C	70 °C
	SE (x 10^{-4})			
Approximation of diffusion	1.4	0.5	1.9	6.5
Two terms	1.1	7.6	1.8	0.7
Two-term Exponential	1.7	8.1	2.4	5.6
Henderson & Pabis	1.8	7.6	2.5	5.4
Mod. Henderson & Pabis	2.0	0.2	198	1.2
Logarithmic	1.0	3.3	1.8	0.6
Newton	2.6	8.1	2.7	4.9
Page	1.7	4.0	2.5	3.0
Thompson	1.6	2.1	2.4	5.6
Verma	1.4	0.5	1.8	0.6
Wang & Singh	43.5	252.3	72.2	5.2
Midilli	1.0	3.3	1.7	0.7
Dryceleaves	113.9	6.5	5.39	0.8

Table 4. Akaike's Information Criteria (AIC) and Schwarz's Bayesian Information Criteria (BIC) of mathematical models fitted to the drying of *C. pachystachya* leaves at air temperature of 40 °C.

Models							
Approximation of diffusion		Two terms		Logarithmic		Verma	
AIC	BIC	AIC	BIC	AIC	BIC	AIC	BIC
-209.04	-202.28	-218.88	-210.43	-220.66	-213.90	-209.05	-202.30

Table 5. Coefficients of mathematical models fitted to the drying of *C. pachystachya* leaves under different temperature conditions.

T (°C)	Models	Coefficients
40	Logarithmic	a = 0.9505**; k = 0.1718**; b = 0.0251**
50	Modified Henderson & Pabis	a = -389.3411**; k = 0.4207**; b = 189.3947**; d = 0.4045**; c = 200.9571**; e = 0.4374**
60	Logarithmic	a = 0.9505**; k = 0.1719**; b = 0.0252**
70	Dryceleaves	a = -0.2486**; b = 0.0147**; c = 1.2344**

**Significant difference at 0.01 probability level by t-test; T = Drying air temperature.

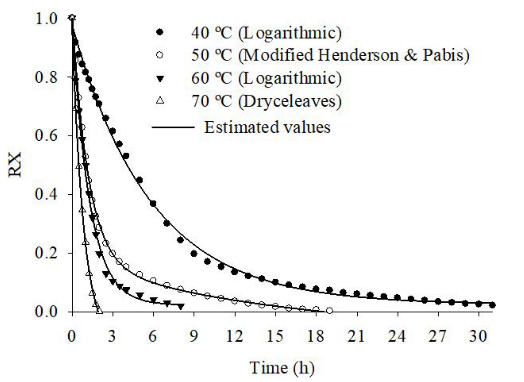

Figure 1. Drying curves experimentally obtained by Logarithm, Modified Henderson & Pabis, Logarithm and Dryceleaves models at air drying temperatures of 40, 50, 60 and 70 °C, respectively.

according to Figure 1 and their respective coefficients are presented in Table 5. It is possible to observe that the chosen models had excellent adjustments with data observed in all drying air temperatures; it was also observed that as the drying temperature increased, the water removal rate also increased, which resulted in shorter drying times.

The diffusion coefficient (D) serves as an indicator of the speed with which water is removed from a product (Silva et al., 2017b), which can be influenced by the increase in drying air temperature (Smaniotto et al., 2017), and results in reduction of water viscosity, facilitating its removal from the capillaries of leaves.

An increase of D was observed as the drying air temperature increased during the drying of *C. pachystachya* leaves. Increments in D increased with increasing of air temperature (Figure 2A), corroborating several studies that have reported the same behavior with increase in drying air temperature (Rodríguez et al., 2014; Dai et al., 2015; Silva et al., 2015; Akpinar & Toraman, 2016; Mghazli et al., 2017). Figure 2B presents the relationship between effective diffusivity and temperature, expressed by the Arrhenius equation.

Mghazli et al. (2017) in the drying of rosemary leaves found D variation from 2.55×10^{-11} to 1.51×10^{-10} $m^2\ s^{-1}$, whereas in mint leaves, values ranged from 0.91×10^{-11} to 10.41×10^{-11} $m^2\ s^{-1}$ (Motevali et al., 2016) and in lemon from 2.61×10^{-11} to 9.24×10^{-11} $m^2\ s^{-1}$ (Tasirin et al., 2014). These results demonstrate effective diffusion coefficient values higher than those found in the present study, showing that *C. pachystachya* leaves have higher resistance to water loss from their inside to the drying air, compared to mint, rosemary and lemon leaves.

Such resistance is probably caused by the higher rigidity and thickness of *C. pachystachya* leaves, but Silva et al. (2017a) highlight the importance of also considering the chemical composition as a factor that influences diffusivity.

The activation energy is the minimum energy value required for the diffusion process to occur (Camicia et al., 2015) and its different values in various products can be attributed to their physical and biological characteristics (Martins et al., 2015). The activation energy for the drying of *C. pachystachya* leaves was

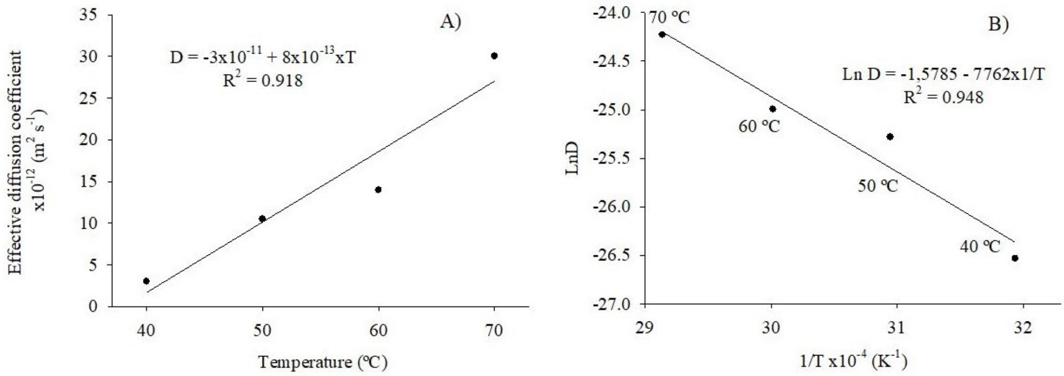

Figure 2. Effective diffusion coefficient as a function of drying temperature (A) and Arrhenius representation as a function of air temperature (B), obtained during the drying of *C. pachystachya* leaves.

64.53 kJ mol^{-1}, which is within the study temperature range, a result close to 63.17 kJ mol^{-1}, found for mint leaves (Motevali et al., 2016), and 63.47 kJ mol^{-1}, found for lemongrass (Martinazzo et al., 2007). In summary, all these results are important for understanding the drying process of C. *pachystachya* leaves in order to guarantee storage and processing in a safe way.

4. CONCLUSION

Among the models analyzed, the Logarithmic model best represented the drying kinetics at temperatures of 40 and 60 °C in the drying of *Cecropia pachystachya* leaves, whereas Modified Henderson & Pabis and Dryceleaves represented temperatures of 50 and 70 °C, respectively. The effective diffusion coefficient increased with increasing air temperature with increments of 8.0 $x10^{-13}$ m^2 s^{-1} for every 10 °C, and the activation energy for liquid diffusion in the drying process was 64.53 kJ mol^{-1}.

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